Lessons Learned in Development and Implementation of the Radionuclide Assessment System

Executive Summary

The Radionuclide Assessment System (RAS) was specifically developed to monitor gamma radiation in existing boreholes at Hanford single shell tank farms. In the course of development, a number of lessons were learned.

- A conventional pickup truck is a poor choice for a logging vehicle. Operator comfort and ergonomic issues are important considerations in the overall system design. Future logging systems should be mounted in a van or crew cab pickup.
- A heavy-duty winch is required for better stability and depth control.
- A wider measurement range is required. The original design criteria called for the ability to quantify ¹³⁷Cs up to 50,000 pCi/g. Concentrations in excess of 100,000,000 pCi/g were encountered during the baseline characterization effort. Future logging systems should be capable of measurement to at least 1,000,000,000 pCi/g ¹³⁷Cs
- The original design requirements assumed that ¹³⁷Cs would be the dominant gamma-emitting radionuclide present in the vadose zone. Other radionculdes, particularly ⁶⁰Co, europium, and processed uranium were also found in significant concentrations.
- Data analysis to determine concentration by application of spectral stripping methods was found to be impractical because of the variety of radionculides encountered, and the inability to compensate for effects of each man-made radionuclide on background count rates for other radionculides.
- Since radionuclide identification and concentration are known from the baseline characterization data, changes in gamma activity can be used to detect on-going migration. Therefore a simpler data evaluation approach based on detection of significant changes between successive logs can be applied. This uses comparison of count rates in spectral windows, rather than detailed evaluation of gamma energy spectra. (A window count is the sum of the counts in a set of contiguous multichannel analyzer (MCA) channels that span a specific energy range.)
- Monitoring measurements should be made "move-stop-acquire" mode. Holding the detector stationary during measurements will improve counting statistics, making it possible to detect more subtle changes.
- The logging system should be based on modification of an existing commercially available mineral, geotechnical or environmental logging system. DOE-GJO / MACTEC-ERS personnel should work closely with the selected vendor to adapt an existing logging system.
- A survey of available gamma ray detectors should be undertaken to identify suitable detector systems for various concentration ranges likely to be encountered at Hanford. Performance criteria include ¹³⁷Cs concentrations between 10⁻¹ and 10⁹ pCi/g, with borehole temperatures as high as 180 to 200 °F

(82 to 93 °C) Electronic compensation systems, such as "pileup rejection," for high count rate effects would also be desirable. It is likely that a combination of detector systems will be required to achieve the required range in measurement capability.

 More attention should be given to gain stability. Magnetic and temperature effects may be significant, and should be addressed in detector design. Other effects, such as peak spreading and dead-time should also be evaluated carefully for each detector system.

Although most of the single shell tanks are considered stabilized, and short-lived radionuclides such as ¹⁰⁶Ru have decayed below detectable levels, the baseline characterization data indicate that many subsurface contaminant plumes with significant gamma activity remain. Both the observed distribution of gamma-emitting contaminants in the subsurface and independent evaluation of groundwater contamination suggest that contaminants associated with tank waste may have reached groundwater. Monitoring is an important component of future remediation activities, both to detect ongoing contaminant migration and to demonstrate stability where movement has stopped.

The lessons learned have resulted in specific recommendations for development and implementation of future monitoring systems:

- Conduct a review of available gamma detectors to identify detector system(s), which can be integrated into logging to provide a measurement range equivalent to 10⁻¹ to 10⁹ pCi/g ¹³⁷Cs under borehole conditions of steel casing, limited diameter, high temperature, and varying magnetic fields.
- Investigate procurement of conventional "off the shelf" logging systems that can be modified for monitoring purposes and work with the vendor to adapt the system to detector(s) identified above. Issues such as vehicle weight, support requirements, depth control, operator ergonomics, data collection system, software, and operational considerations would be addressed.
- Adapt a "move-stop-acquire" logging mode instead of continuous logging to improve counting statistics and thereby facilitate identification of subtle changes in subsurface radioactivity profiles.

1. Introduction

Since 1995, the U.S. Department of Energy Grand Junction Office (DOE-GJO) has performed spectral gamma logging of existing boreholes in the vadose zone in the vicinity of the Hanford single-shell tanks. High resolution spectral data have been used to determine the current nature and extent of gamma-emitting contamination in the vicinity of the Hanford single shell tanks. This data set provides a baseline to which previous geophysical logs and future monitoring data can be compared to identify and assess contaminant migration or stability. With the completion of the baseline characterization project, the U.S. Department of Energy Office of River Protection

(DOE-ORP) has requested that DOE-GJO develop and implement a monitoring program in selected boreholes in the single-shell tank farms.

Prior to 1994, vadose zone monitoring was performed by Tank Farms personnel, using Geiger-Mueller (GM) or scintillation detectors. These systems recorded total gamma count rate as a function of depth. Data are available in electronic format from 1975 to 1994, and independent evaluation of these logs by others (e.g. Randall & Price, 1998) has confirmed the presence of subsurface contaminant plumes associated with tank farm operations and also provides indications of continued contaminant movement.

By the early 1990's, however, a number of serious deficiencies in the borehole surveillance program had been identified. These included (DOE-1995):

- The boreholes are not spaced closely enough around each tank to ensure detection of a leak
- The logging system did not differentiate gamma rays by energy level and was unable to identify radionculides from characteristic gamma emissions
- Logging speed and sampling intervals were inappropriate for the detectors in use and for the depth distribution of the contaminants.
- No dead-time correction.
- Inappropriate criteria for leak detection and monitoring by gamma logging.
- Inappropriate detector calibration based on point-source exposure rate standards.
- Inadequate spatial resolution as a result of large depth intervals between readings.
- The detectors were paralyzable and may not have provided accurate measurements in zones with very high gamma count rates.

In 1994, gross gamma logging was discontinued in tank farm boreholes, and plans were made to develop and implement a new monitoring system. The spectral gamma logging system (SGLS) used to acquire the baseline data set provided high-quality data, but it depended on high-purity germanium (HPGe) detectors, which are too complex and difficult to operate for routine monitoring. A simpler logging system based on thallium-activated sodium iodide (NaI(Tl)) detectors would be easier to operate and capable of faster logging speeds. This would provide more cost-effective monitoring data. The new system was originally referred to as the Leak Verification Monitoring System (LVMS), but later renamed the Radionuclide Assessment System (RAS). Development of this system began in 1995. Components, including a vehicle, winch and cable, and three NaI(Tl) detectors, were procured and a monitoring system was assembled. However, development was suspended because of funding issues. In FY2001, funding was provided to complete development of the RAS and to begin monitoring in tank farms boreholes. By July, 2001, the RAS was operational as a monitoring system.

The purpose of this document is to provide a discussion of problems and issues encountered in development and implementation of the RAS, and to describe how these problems were addressed. Individual sections will address specific components of the RAS, as well as issues associated with calibration, operation, and data evaluation. These "lessons learned" can be applied to development and implementation of future logging

and monitoring systems for tank farms and other waste sites at Hanford. Many of the lessons learned can also be applied to spectral gamma logging and monitoring operations at other DOE sites

2 Design Requirements

The SGLS used for the baseline characterization provide very high quality data, but the high-purity germanium (HPGe) detectors require liquid nitrogen cooling and relatively long count times. Moreover, data analysis and interpretation is a relatively complex process. Support requirements, data collection rate, and analytical effort argue against use of the SGLS for routine monitoring purposes. The RAS was conceived as a simple spectral gamma logging system that could be easily maneuvered inside tank farms and operated by Tank Farms personnel. Use of simpler NaI(Tl) detectors reduces support requirements and logging complexity and increases the data collection rate, albeit at the expense of energy resolution. Specific design requirements for the RAS include the following:

- Rapid deployment and ease of operation.
- Spectral gamma measurement capability
- High efficiency for good statistical precision at relatively fast logging speeds
- Detect and assay background gamma radiation associated with natural potassium (⁴⁰K), uranium (²³⁸U) and thorium (²³²Th) "with a precision that permits contaminant detection to the minimum concentrations specified in applicable regulations."
- Detect ¹³⁷Cs and ⁹⁰Sr
- Maximum contaminant level of 50,000 pCi/g (¹³⁷Cs)
- High degree of repeatability

⁹⁰Sr is a "pure" beta-emitting radionuclide: there is no gamma ray associated with the decay of ⁹⁰Sr and it is impossible to directly detect ⁹⁰Sr inside a steel casing, since the metal effectively shields beta particles. However, when high-energy beta particles interact with the steel casing, incoherent low-energy (<350 KeV) gamma rays known as brehmsstrahlung are generated, which can be detected inside the casing. With the SGLS, the presence of ⁹⁰Sr is inferred through spectral shape factor analysis, or when anomalous low-energy (< 350 KeV) gamma counts are observed with no specific peaks at energies characteristic of man-made gamma emitting radionuclides. With the RAS, the presence of ⁹⁰Sr is indicated by the presence of anomalous gamma counts, particularly in the spectral region below 350 KeV.

The initial design criteria were developed during the early stages of the baseline characterization project when ¹³⁷Cs was considered to be the primary target. Subsequent evaluation of baseline data indicated that man-made radionuclides other than ¹³⁷Cs are also of concern.. These include ⁶⁰Co, uranium (²³⁵U and ²³⁸U), europium (¹⁵²Eu and ¹⁵⁴Eu), ¹²⁵Sb, and ¹²⁶Sn. Since the baseline data set provides the identity and concentration of each radionculide within a borehole interval, it is more important that the RAS be able to detect *changes* in gamma activity between successive runs, rather

than identify and quantify specific radionuclides. Although gamma spectra are collected for evaluation if necessary, routine analysis is based on comparison of count rates between successive runs

Since counts are compared directly, additional uncertainties associated with the concentration calculations, such as casing correction or calibration error, are eliminated, making it easier to distinguish subtle changes.

Finally, results from the high rate logging system (HRLS) indicate that ¹³⁷Cs concentrations in excess of 100,000,000 pCi/g may be present.

3 System Components

The following is a brief description of the RAS and its individual components. More thorough descriptions are found in the *RAS Operational Test Plan Results* (DOE 2001), in the *RAS Logging Procedures* (DOE 2001), and in the *RAS Preventative Maintenance Plan* (DOE 2001).

The RAS monitoring system is mounted in a 1996 Chevrolet diesel pick-up. A DC to AC power inverter supplies 120 volt AC power for the logging and data collection system. A Mount Sopris winch moves the logging sonde in a borehole. The winch utilizes 500 ft of 0.25-in diameter, 7-conductor logging cable. A winch controller mounted in the cab of the pick-up allows the operator to control both the speed and spooling direction of the winch. The logging sonde consists of a telemetry section and a detector section. The telemetry section utilizes an Ortec Micro NOMAD MCA to collect and transmit spectra. Each detector section contains a NaI(Tl) crystal coupled to a photo-multiplier (PM) tube which is powered by an Ortec PM tube base and power supply. Three interchangeable detectors were developed to provide a wide measurement range. The primary difference between the detectors is the size (efficiency) of the sodium iodide crystal. A laptop computer is used to record the spectra, extract the window counts from each spectrum and record the depth at which each spectrum is acquired.

Nearly all RAS components were procured and assembled in 1996. When funding resumed in FY2001, there was little opportunity to make major changes in system components to take advantage of information and experience gained in the baseline characterization program. There are a number of alternative system components that could have been selected to make logging operations in the Hanford tank farms more efficient and effective. These are discussed in the following sections.

3.1 Vehicle

A 1996 Chevrolet diesel pickup was procured through GSA in Grand Junction, Colorado for use as the RAS. At the time, a van had been requested, but the pickup was the only vehicle available. The diesel engine is better suited to logging operations than a gasoline

engine, since the truck is required to run at idle speed for long periods while boreholes are logged. The diesel provides more power at idle speed to support the alternator and inverter, and is less likely to overheat than a gasoline engine. However, the use of a pickup for the logging system has resulted in several problems, which will be discussed in detail below.

The logging system is operated from the passenger seat in the pickup. Since the operator must be able to observe the borehole and logging system during monitoring operations, it was necessary to reverse the passenger seat. Originally, the passenger seat was mounted on a swivel and turned to face the rear of the truck. The logging computer was located on a box that rested between the driver's seat and the passenger seat. This configuration was extremely awkward and uncomfortable because the operator had to turn sideways in the seat to operate the computer. This arrangement was changed so that the computer was mounted on a tray at the back wall of the cab. This modification allows the operator to work with the computer while facing the logging operation.

There is very little legroom with the seat facing backward. Moving the seat back (forward) as far as possible resulted in a crack in the windshield where the seat back contacted the windshield. The backrest was removed so that the seat could be moved farther back, but this resulted in less headroom for the operator because of the slope of the windshield. It has been suggested that a hole be cut into the back wall of the cab so that the operator's legs can extend into the pickup bed. While this may alleviate the cramped legroom, it will compromise the integrity of the vehicle and may result in leakage during bad weather. Operator comfort is an important consideration, because several hours are required to log most borehole intervals. Any future logging system should be installed in a van, or at least in a pickup truck with a crew or extended cab. Another possibility would be to install an enclosed operator's station in the rear of the vehicle.

Glare from ambient sunshine made it difficult to see the original laptop computer screen. This was addressed in two ways. First, a new laptop computer that used an active matrix screen was purchased. Second, the windows of the pickup were tinted. Glare can be a major problem because the logging system and data collection are controlled from the computer. It is not always possible for the operator to orient the vehicle to minimize glare. An enclosed operator station in the back of a van or in the rear of a pickup would be a more effective solution for glare.

There is a significant amount of wasted space in the bed of the pickup. A canopy is necessary to protect the logging equipment from the weather, but this restricts access to most of the space between the logging equipment at the rear of the pickup and the cab. Two pieces of equipment mounted in this space are the power inverter and the field verifier. There is very little need to access the power inverter unless repairs are required, but the field verifier needs to be accessed by the health physics technicians on a regular basis for source integrity tests and dose rate measurements. The only way to reach the verifier with the present equipment configuration is to climb over the winch.

Overall, a conventional pickup with a canopy is a poor choice for a logging vehicle. Future logging systems should be mounted in a van or a crew cab pickup. If a conventional pickup is to be used, the pickup bed should be removed and a custom body installed on the truck chassis. This would allow room for an enclosed operator station with adequate headroom and legroom, as well as better arrangement of the logging system and support components.

3.2 Logging System

The logging system consists of the winch, logging cable and winch controller used to move the sonde up and down the borehole and to transmit data signals from the sonde to the data collection system. A mast assembly positions the logging sonde over the borehole.

The original logging system had to be modified because of several deficiencies, which are discussed below.

Winch

A Mount Sopris MX series winch was originally installed in the RAS. This was a light-weight winch that used a 1/8 inch diameter single conductor logging cable. This winch had difficulties holding the sonde at a constant depth. After several attempts to correct this problem, it was determined that the weights of the sondes were near the maximum weight rating for this winch. This winch was replaced with a heavier Mount Sopris MN series winch and new winch controller.

Cable

The single-conductor logging cable complicated the computer telemetry interface because the data signal had to be carried (duplexed) on the same conductor as the downhole power. The small diameter logging cable was also very easily kinked. The new winch is equipped with ¼ in diameter seven-conductor logging cable. The new logging cable allows the data signal and power to be run on separate conductors. This greatly simplifies the telemetry and makes the data collection system more robust. The larger diameter cable is also more resistant to kinking.

Winch Controller

The original Mount Sopris MX Series winch was controlled from two remote controllers connected to the main control console mounted on the side of the winch. One of these controllers was permanently mounted on the rear wall of the truck cab, and the other was a pendant (wired remote) that could be operated at a distance of up to 10 ft from the rear of the truck. These two controllers were built in Grand Junction and were not part of the original equipment purchased from Mount Sopris. To compensate for the weight of the sonde, a slight upward speed had to be applied to the winch motor to hold the sonde

stationary in the borehole. The controllers also suffered from electrical shorts and were not compatible with the new winch.

The new Mount Sopris MN Series winch included a new controller, which is more dependable than that on the previous winch. The original RAS logging system did not include a sensor that could shut off the winch when cable tension fell outside maximum or minimum values. This is an important feature that prevents damage to the system should the sonde become stuck in the borehole or in the top sheave of the mast. The new MN series winch is equipped with a tension sensor and monitor, which allows minimum and maximum tension limits to be set. The winch is automatically shut off when these limits are exceeded.

The only drawback to the new winch controller is size. It was designed to mount in a standard instrument rack and the mounting had to be revised to fit inside the RAS vehicle cab. The controller was installed vertically inside a box that was mounted between the two seats in the cab of the truck.

Mast vs Boom

The RAS utilizes a mast and base plate assembly instead of a boom to position the logging sonde over the borehole. The original RAS used just one base plate, the 6 in diameter. This was exactly 6 inches OD and would not fit in most 6 in ID boreholes, so a new base plate with 5 ¾ in OD was made. A 3 ¾ in OD base plate was also made for use in the 4 in ID boreholes. Future logging systems should be capable of using either a mast or a boom, depending on borehole access.

Logging Mode

For monitoring purposes, logging speed and depth control are important issues. Since measurements are made by counting for a time interval while the sonde is moving in the borehole, the character of the log and the vertical range over which values are averaged depends on the interrelation between logging speed and counting time. In general, longer counting times provide better statistics, but require slow logging speed to achieve the same depth resolution. Data repeatability can be improved by operating the monitoring system in "move-stop-acquire" mode, where the sonde is held stationary for each measurement and then moved to the next depth increment.

Logging System Recommendations

In its present configuration, the logging system is adequate for monitoring purposes. Future monitoring systems should use heavy-duty winches. Depth control requirements for a monitoring system are more stringent than those for conventional logging systems. Tension limit switches are also important to prevent damage to the sonde, cable or cable head if the sonde becomes stuck in the hole or hits the sheave wheel.

3.4 Logging Sonde

The RAS sonde consists of two sections. The upper section contains a multichannel analyzer and the telemetry components. Any one of three detector modules can be connected to this section. A number of problems and issues were encountered with the logging sonde. These included the connection between the telemetry section and the detector modules, the measurement range of the detectors, gain shift, MCA/telemetry, detector housing diameters, and the borehole environment.

Module Connections

The connection between the telemetry section and the detector modules uses a pin to slip ring system. As the detector module is threaded into the telemetry section, pins on the detector module contact slip rings on the telemetry module. The slip rings are recessed inside the telemetry module while the pins are exposed at the end of the male thread connection on the detector module. This creates a potential for the pins to be broken or bent. A better design would place the pins inside the female thread on the telemetry module and the slip rings on the male thread detector sections.

Detector Range

Original design criteria required the RAS to be capable of detecting ¹³⁷Cs from background levels up to concentrations of about 50,000 pCi/g. This range of activity proved to be too wide for a single detector. Three detectors with overlapping ranges were purchased:

RAS NaI(Tl) Detectors

Detector	Dimensions (diameter by	Approximate Measurement Range (pCi/g Cs-137)		
	length, in inches)	Minimum	Maximum	
Large	3 by 12	Background	10 ³ pCi/g	
Medium	1.5 by 2	10 pCi/g	10 ⁴ pCi/g	
Small	1 by 1	100 pCi/g	10 ⁵ pCi/g	

Although the small detector is capable of measurements at about twice the maximum level of 50,000 pCi/g specified in the original design requirements, actual concentrations greater than 100,000,000 pCi/g have been encountered in tank farms boreholes. Therefore, additional detectors and/or shielding will be required to provide the full measurement range.

Gain Shift

Gain refers to the amplifier setting which controls how each pulse is correlated to a MCA channel number. In the detector, each gamma photon produces an electrical pulse whose

height (voltage) is proportional to the energy of the gamma ray. In logging practice, gain is adjusted so that counts associated with a particular pulse height are tallied in a specified MCA channel. Over time, the gain may change slightly, so that those counts may be assigned to a different channel. Gain shift may also occur as the result of external influences, such as temperature or magnetic fields. Minor shifts in gain result in the appearance of peak spreading when pulses are shifted to adjacent channels, while major gain shifts may distort peaks to such an extent that the peaks are no longer correctly recognized. Stable gain is especially important where gamma peaks are used to identify radionuclides or where counts in spectral windows are to be compared.

Temperature effects on radiation detectors are difficult to avoid in logging practice. When logging in winter or summer, temperature variations on the order of 50 degrees Fahrenheit are possible, simply from differences in ambient air temperature and the subsurface temperature related to the normal geothermal gradient. In tank farms, thermal anomalies may be associated with intervals of intense contamination.

Magnetic fields can affect electron currents in photomultiplier tubes, which will be expressed as a gain shift. The carbon steel casing used in tank farms boreholes tends to have detectable magnetic anomalies, particularly at welded joints. When the detectors were originally fabricated, this phenomenon was overlooked and inadequate magnetic shielding was used in the detectors. If future logging systems use scintillation detectors, the photomultiplier tubes should be adequately shielded against magnetic fields.

Some detector systems use gain stabilization to help control drift. This requires that the spectra always have a recognizable peak present, which the stabilization software can use to make continuous gain adjustments. This can be accomplished by inclusion of a small radioactive source or flashing light near the detector. Obviously, the counts associated with this source add background counts to the spectrum, which must be accounted for during analysis. The current RAS system does not have gain stabilization capability. Consideration should be given to using gain stabilization in future logging systems.

MCA/Telemetry

The RAS logging system uses an Ortec MicroNOMAD MCA, which has been repackaged into the telemetry module. This MCA has a tendency to lock up during logging, requiring a system reset. Lock-up tends to occur at high count rates, and appears to be a characteristic of the system. In future logging systems, a more robust counting system should be used to avoid lock ups. Also, the concept of a downhole MCA should be reconsidered. Moving the MCA uphole would simplfy the downhole electronics, possibly eliminating the need for a separate telemetry section.

Detector Diameter

The large detector housing has an outside diameter of 4 inches, while the medium and small detector housing have an outer diameter of 3 inches. Although the RAS is designed

to log boreholes with diameters as small as 4 inches, it is difficult to measure low levels in 4-inch boreholes, since the medium detector must be used.

Borehole Environment

Elevated temperatures are known to exist in some boreholes as a result of high levels of radioactive decay activity. Temperature logging in SX and A tank farms has measured borehole temperatures in excess of 160 °F. Discussions with the manufacturer of the detectors (Alpha Spectra) revealed that the detectors fabricated for the RAS are only rated to 95 °F. The RAS PM tubes are only rated to 140 °F, and the cathode material inside the tubes begins to degrade at 194 °F.

Detector Recommendations

From the above discussion on detectors, it is apparent that at least one, and probably two additional detectors will be required to increase the measurement range to concentrations as high as 10⁸ pCi/g. Since the high temperatures occur in zones of high radioactivity, these detectors should be designed to function at much higher temperatures. Tentatively, 200 °F is suggested as the minimum operating temperature for high rate detectors.

The baseline characterization data provides a definition of the required measurement range for future monitoring systems, and extremes of borehole conditions in the vadose zone are known. Given such a wide range of borehole conditions and gamma flux, it is doubtful that any single detector type is ideal over the entire range. A thorough evaluation of existing gamma ray detectors should be performed to provide a basis for detector selection. Detector characteristics that are advantageous in low levels of radioactivity such as efficiency and stopping power, can become a liability in high levels of radioactivity. Factors to be considered include measurement range, size, support requirements, operating requirements, and environmental restrictions. This evaluation would be carried out by a review of publications and vendor literature, supplemented by limited testing.

3.5 Data Collection System

The data collection system includes the downhole MCA/telemetry unit, the winch controller, the depth encoder, and the laptop computer. Count data are collected in the MCA and the energy spectra are transmitted up the cable via the telemetry link. A serial (RS-232) interface transfers data from the uphole telemetry unit located in the detector power supply to the laptop computer. Depth information is transmitted to the computer from the depth encoder via an RS-232 interface. During logging, spectra are transmitted from the MCA and combined with depth information from the depth encoder. The software records counts in each of eight spectral windows, as well as total counts as a function of depth. At the end of the log run, the count data, as well as spectra files, header data and verification spectra are transferred to a 250-MB ZIP disk via a universal

serial bus (USB). The ZIP disk allows monitoring data to be transferred to hard disk on the MACTEC network on a daily basis.

The existing data collection was originally set up in 1996, and modified to include the ZIP disk in 2001. In future systems, consideration should be given to replacing the serial interfaces with USB, or by using a card to collect data directly. The card could be mounted in a laptop docking station, or a rack-mounted computer could be provided if sufficient space is available.

Data Collection System Recommendations

If a conventional "off the shelf" logging system is procured as recommended, it will have a data collection and storage system developed by the vendor. Modification of this system to meet monitoring requirements is likely to be the most cost-effective option.

3.6 Operating Software

The software utilized to operate the data collection system (LVMON) was developed by MACTEC-ERS. The software stores the gamma spectra and records total counts and counts for each of eight spectral windows as a function of depth. Between 1996, and 2001, there were significant advances in computer technology. An upgrade of the laptop computer and conversion of the operating system to Windows 98 required that the original software be re-written to function in a 32-bit computing environment. As discussed above, the system experiences problems with MCA lockup at high count rates and a slow depth refresh rate. Both of these problems appear to be hardware issues. Otherwise, the software functions correctly. The primary output of the logging system is a text file containing each of the window counts, total counts, live time, and dead time as a function of depth. This file can be directly imported to Microsoft EXCEL® for data analysis.

Operating Software Recommendations

If a conventional "off the shelf" logging system is procured as recommended, it will have operating software developed by the vendor. Modification of this software to meet monitoring requirements is likely to be the most cost-effective option.

4. Calibration

Since the baseline characterization data provide radionuclide identification and initial concentration, the primary function of the RAS is to detect changes in gamma activity over time. Because calculation of concentrations is not the primary objective, it is not necessary to calibrate the RAS in the usual sense. That is, no correlation between instrument response and concentration has been derived. Initially, it was thought that the system could be calibrated to determine ¹³⁷Cs concentration by subtracting or "stripping"

the contribution of naturally occurring potassium-40 (⁴⁰K), uranium-238 (²³⁸U), and thorium-232 (²³²Th) from a ¹³⁷Cs window that would be defined to capture counts due to the 661.6-keV 137Cs gamma ray. Coefficients for the stripping calculations could be determined from measurements in the GJO and Hanford calibration models. However, the presence of other man-made radionculides such as cobalt-60 (⁶⁰Co) and europium 152 and –154 (^{152/154}Eu) cannot be accounted for in the stripping process, unless calibration models are constructed to isolate the effects of each radionuclide on background levels for the others. Therefore, the decision was made to compare counts in pre-defined spectral windows to assess changes in activity. Measurements have been made at GJO and in the Hanford calibration models to assess the performance of the RAS detectors in known radiation environments and to determine measurement precision.

4.1 Initial Calibration

Calibration measurements were made with the RAS in the GJO calibration models in FY1996 and at the Hanford calibration models in FY2001. Data from these measurements were used to determine system characteristics. For example, the system dead time effect was investigated and found to be negligible, and the overall measurement precision was determined to be suitable for monitoring purposes. Results of the initial calibration are discussed in detail in *Initial Calibration of the Radionuclide Assessment System* (Koizumi, 2001).

4.2 Verification Measurements

Verification measurements are made to assess the day-to-day performance of the logging system. A portable, sealed potassium-uranium-thorium (KUTh) source was acquired for field verification measurements. This source contains potassium, uranium and thorium compounds. Activities of decay progeny in the uranium and thorium series are presumably in secular equilibrium with the parent radionuclides. This represents a relatively stable source and daily measurements made with this source can be used to assess detector performance over time.

5. Operational Issues

Hanford Tank Farm personnel operate the RAS system, and two operators are assigned to operate the system on a daily basis. Six operators have been training on the system and have satisfied a qualification requirement of the tank farms contractor CH2M Hill Hanford Group (CHG). A meeting was held with all six operators to collect their input on operational issues of working with the RAS. The discussions below summarize their suggestions.

The main complaint was the vehicle itself and in particular the limited legroom for the operator. They suggested using a full-size (3/4 ton) diesel van. The van should be

equipped with swiveling cloth captain's chairs so both operators can view the computer screen. The gross vehicle weight should be kept under 10,000 lbs to avoid dome loading issues. Other options suggested by the operators were that the van should have tinted windows, a protective plate for the fuel tank and a block heater.

The operators liked the current winch installed on the RAS but would like to have the following modifications: 1) a remote winch controller at the rear of the vehicle, 2) computer control and, 3) faster speed for retrieving the sonde from the borehole. They also like the idea of using a boom instead of the mast assembly. The mast assembly would still have to be used (in combination with the boom) for inaccessible boreholes.

The logging sonde should be made as light as possible, preferably less than 40 lbs. The operators would also like to eliminate the telemetry section all together.

Their main suggestion concerning the computer and software was to have the computer control the winch, and that the computer be equipped with a cordless or trackball mouse. Another suggestion was to change the software so all readouts were near the same location on the screen, and revise the software so that data transfer to the ZIP drive could be completed once per day.

Other suggestions that were offered during the meeting included the following: 1) install a base radio in the truck, 2) use a larger engine hour meter, 3) install a shore power hookup, 4) utilize better designed, more ergonomically efficient equipment storage, 5) better access to the KUTh verifier, 6) extra 110 V ac outlets and, 7) external flood lights.

6. Data Analysis

The development of a data analysis methodology for the RAS is discussed in detail in Appendix A. Originally, the intent was to calculate ¹³⁷Cs concentrations by stripping or subtracting the contribution from naturally occurring radionculides associated with ⁴⁰K, and the ²³⁸U and ²³²Th decay series. This is possible when only one target radionuclide is present, but becomes impossible when a variety of man-made radionuclides may be present. Energy resolution capability of NaI(Tl) detectors is not adequate to isolate specific energy lines. Measurement is based on counts recorded in relatively broad spectral windows. The background counts in each window due to other radionuclides must be subtracted to determine the counts associated with the target radionuclide. The existing calibration models are adequate to determine the effects of naturally occurring radionuclides on background counts in any spectral window, but calibration would require an additional series of models to determine the effects of each target radionuclide on background counts in each spectral window. Since both radionuclide identity and concentrations are known from the baseline characterization data, it is not necessary to determine concentrations with the RAS. A simplified data analysis approach oriented toward detecting changes in subsurface radioactivity levels is more appropriate.

The gamma spectra recorded by the RAS is subdivided into eight contiguous windows and count rates are recorded for each window. The rationale behind the definition of each window is discussed in Appendix A.

To a first approximation, changes in contamination profiles can be identified by simply comparing plots of successive log runs. When necessary, count rates can be corrected for decay using radionuclide identification from the baseline data. Areas of possible contaminant migration can be identified by changes in count rate over a depth interval or changes in the depth over which anomalous activity occurs.

Like all radiation measurements, RAS data are subject to random fluctuations associated with the radioactive decay process. Therefore, it will be necessary to determine if observed differences in count rates are statistically significant. This follows a method described in Knoll (2000), in which limits are established at a pre-determined level of significance. One limit defines the level at which there is no statistically significant difference in count rates, and the second limit defines the level at which a statistically significant difference in count rates exists. The mathematical derivation of these limits is discussed in Appendix A.

Data Analysis Recommendations

The current graphical scheme of data analysis is relatively simple and quickly identifies changes in radioactivity levels. This allows data interpretation to be carried out quickly after logging is completed. When necessary, gamma spectra can be examined for more information, and the SGLS or HRLS can be used if more precise measurements are required.

The data analysis approach for future logging systems will be based on the most effective method for the specific detector system(s).

7. Conclusions

The RAS was successfully completed and deployed to conduct monitoring operations in boreholes surrounding the Hanford single shell tanks. Although a number of problems were encountered, the RAS has proven useful in detecting intervals of potential contaminant movement in the vadose zone. The data can be quickly plotted for visual analysis or a more detailed statistical analysis can be performed as necessary.

Even though the SSTs are being stabilized and tank contents are being transferred to double shell tanks, contaminant plumes exist in the subsurface, and a monitoring program is necessary to detect any continuing migration. Although short half-life radionuclides such as ¹⁰⁶Ru have decayed below detectable levels, and other radionuclides that constitute the greatest risk, such as ⁹⁰Sr or ⁹⁹Tc, cannot be detected directly with gamma measurements in cased holes, the baseline data and monitoring experience to date indicate that significant levels of gamma activity remain in the vadose zone. Results of

the baseline characterization and independent evaluation of groundwater data indicate that contaminant plumes from SST leaks may have impacted groundwater. The mobility of these contaminant plumes is an important factor in assessing the ultimate risk to human health and the environment, and in selecting and implementing appropriate remedies. Where contaminant plumes are shown to be stable, consideration can be given to leaving the material in place to attenuate naturally through radioactive decay, with appropriate monitoring. For example, ¹³⁷Cs plumes with concentrations in excess of 10⁸ pCi/g have been detected. Excavation, transport and disposal of soil with these contamination levels will result in a significant radiation dose to remediation workers and represents a potential for airborne contamination. ¹³⁷Cs has a half-life of 30.7 years and seems to be relatively immobile in the subsurface now, even though it was apparently carried great distances in the past by movements of liquids from tank leaks.

8. Recommendations

The baseline characterization, independent evaluation of historical gross gamma data, and assessment or groundwater data all indicate that significant vadose zone contamination exists. Monitoring gamma-emitting contaminants in the vadose zone through existing boreholes is an important and cost-effective component of the overall site remediation effort and should be continued. Monitoring is also important in boreholes around tanks in which retrieval operations are underway, particularly when liquids are being added as part of the retrieval process. At present, only one system is available to support monitoring operations in almost 800 boreholes. Additional systems must be procured to support a reasonable monitoring frequency, as well as to avoid major gaps in the monitoring program that might result from equipment failure. Also, the ongoing vadose zone baseline characterization project has been extended to existing boreholes in and around liquid waste disposal sites in the Hanford 200 Areas. Evidence of subsurface contamination has already been detected in a number of these boreholes, and it is likely that many will require monitoring in the future.

Although the RAS has been effective in monitoring operations for tank farms, lessons learned in its development and implementation have identified a number of shortcomings, which should be corrected in subsequent monitoring systems. Given below are specific recommendations for development and implementation of borehole logging systems for monitoring at the Hanford Site.

• Extend Measurement Capability to 1,000,000,000 pCi/g ¹³⁷Cs

The existing RAS is capable of measurements up to about 100,000 pCi/g. This is about 4 orders of magnitude below the required capability. Additional detectors and shielding will be required to achieve this range.

• Perform a Review of Available Gamma Detectors

The ability to make reliable and repeatable measurements in zone of intense gamma flux is an important requirement for the monitoring system. NaI(Tl) detectors generally work well at low to intermediate count rates, but may not be

the best choice in high rate zones. Detector characteristics that are advantageous at low levels of radioactivity may become liabilities at high activity levels. Environmental factors, such as borehole temperature and magnetic fields associated with steel casing must also be addressed. The detector review would rely on published literature and vendor information to identify and evaluate various detectors and detector configurations suitable for all or part of the anticipated measurement range. The goal would be to identify optimum detectors for specific radiation levels and borehole conditions, and to integrate those detectors within sondes compatible with the new logging system.

• Procure a Conventional Logging System

Several companies, notably Century Geophysical and Mt Sopris Instruments, manufacture integrated logging systems for mineral, geotechnical and environmental applications. These systems are typically mounted in a crew cab pickup or van and provide power supply, winch, winch control, depth encoding and data collection systems that are specifically designed for logging operations. Many of the ergonomic and equipment compatibility issues encountered with the RAS development will already have been addressed. These companies can also provide detectors that can be used at Hanford. For example, conventional spectral gamma and neutron moisture detectors could be used in intervals of low contamination. Many commercially available sondes are designed to be run in combination. This allows gamma counts and neutron moisture data to be collected in a single pass. Some degree of equipment and software modification will likely be required to deal with man-made gamma emitting contaminants, which are not usually encountered outside the DOE environment. This can be best accomplished by working in cooperation with engineers who have experience in development of conventional logging systems. Specific factors to be evaluated as part of the logging system procurement would include:

- Gross vehicle weight and maneuverability
- Support requirements
- Operator ergonomics, equipment access and operational considerations
- Winch stability, depth control and logging speed
- Electrical system
- Data collection system and software
- Compatibility with special-purpose detectors identified above.
- Capability for modification to accommodate special requirements associated with monitoring man-made radionuclides

• Hold the detector stationary for measurements

Baseline data indicate that contaminated intervals frequently occur as very thin zones. Radiation levels can change rapidly over very short depth increments. If the detector is moving as spectra are acquired, then radiation levels may change significantly during the count time. This may affect the counting statistics, making it more difficult to detect subtle changes. When the detector is held stationary, the radiation field is constant during the count time, and response is

more predictable. Also, it is not necessary to maintain stable sonde movement speeds to the same level of precision.

9. References

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Appendix A RAS Data Evaluation

The original approach to analysis of the data acquired with the RAS was based on work of R.D. Wilson and D.C. Stromswald (1981) as part of the National Uranium Resource Evaluation (NURE) program. It used counts in three spectral windows referred to as the K, U, and T windows. The K window collected counts due to the 1460.8 keV peak associated with ⁴⁰K, the T window collected counts due to the 2614.5 keV peak associated with ²³²Th, and the U window collected counts due to the 1764.5 and 2204.1 keV peaks associated with ²³⁸U. Nuclides in the ²³⁸U and ²³²Th decay series give rise to many gamma rays with various energies, so that the counts in each window are a function of all three radionuclides. Potassium, uranium and thorium concentrations were calculated from the matrix equation:

$$\begin{bmatrix}
K_{con} \\
U_{con} \\
T_{con}
\end{bmatrix} = [A]^{-1} \begin{bmatrix}
K_{win} \\
U_{win} \\
T_{win}
\end{bmatrix}$$
(1)

Where K_{win} , U_{win} , and T_{win} are the respective window count rates, and K_{con} , U_{con} , and T_{con} are the concentrations. [A]⁻¹ is a 3 X 3 matrix called the calibration matrix, whose elements are determined from measurements in calibration models where K_{con} , U_{con} , and T_{con} are known. This approach is commonly used in conventional spectral gamma logging, where man-made gamma emitting radionuclides are not expected to be present.

This approach was originally considered because the key potassium, uranium, and thorium gamma rays have higher energies than the ¹³⁷Cs gamma ray (661.6 keV), and ¹³⁷Cs was thought to be by far the predominant constituent in subsurface contamination. Gamma rays from ¹³⁷Cs would not contribute significantly to the counts in the K, U, and T windows, and the count rates in those windows could therefore be used to calculate the potassium, uranium, and thorium concentrations. Using the concentrations, the potassium, uranium, and thorium count rate contributions to a cesium window centered at 661.6 keV could be calculated, and the count rate due to ¹³⁷Cs alone could be inferred. Presumably, the count rate due to ¹³⁷Cs would be proportional to the ¹³⁷Cs concentration.

A fundamental assumption in the NURE approach is that both the uranium and thorium decay series are in secular equilibrium, since the sources of the peaks used are daughter nuclides well down in the decay chain. The 1764.5 and 2204.1 keV gamma rays used for uranium are emitted by ²¹⁴Bi and ²¹⁴Pb, respectively, while the 2614.5 keV peak used for thorium is emitted by ²⁰⁸Tl. Under the assumption of secular equilibrium, the activity of the parents ²³⁸U and ²³²Th can be calculated. The time required for attainment of secular equilibrium is on the order of several million years, so man-made (chemically processed) uranium does not result in elevated gamma activity at these energies. However, the presence of radon may affect uranium window counts. ²²²Rn is a highly mobile gas. ²¹⁴Bi and ²¹⁴Pb are short-term radon daughters, so secular equilibrium with ²²²Rn is

achieved in a matter of hours. The presence of excess ²²²Rn will thus result in anomalous counts in the U window, while the K and T windows are relatively unaffected.

By the time the development of the RAS was being completed, results of the initial baseline characterization in the twelve tank farms indicated that a number of man-made radionuclides in addition to ¹³⁷Cs were present in significant amounts. Table 1 lists major man-made radionuclides encountered during the Hanford Tank Farms Vadose Zone Characterization Project.

Table 1. Man-made radionuclides detected by the Hanford Tank Farms Vadose Zone Characterization Project

vadose Zone Characterization i Toject								
	half life	Primary Ga	Primary Gamma Rays		Secondary Gamma Rays			
radionuclide	years	E, keV	Y, %	E, keV	Y, %			
⁶⁰ Co	5.2714	1332.50	99.98					
		1173.24	99.90					
¹²⁵ Sb	2.7582	427.88	29.60	600.60	17.86			
				635.95	11.31			
				463.37	10.49			
¹²⁶ Sn	1.E+5	414.50	86.00	666.10	86.00			
				694.80	82.56			
¹³⁷ Cs	30.07	661.66	85.10					
¹⁵² Eu	13.542	1408.01	20.87	121.78	28.42			
				344.28	26.58			
				964.13	14.34			
				1112.12	13.54			
				778.90	12.96			
¹⁵⁴ Eu	8.593	1274.44	35.19	123.07	40.79			
				723.31	20.22			
				1004.73	18.01			
				873.19	12.27			
²³⁵ U	7.04E+8	185.72	57.20					
²³⁴ Pa		1001.03	0.84	811.00	0.51			
(man-made ²³⁸ U)				766.36	0.29			

Unfortunately, the data analysis approach described above could only be used to detect and quantify ¹³⁷Cs in the absence of other man-made radionculides. If, for example, ¹⁵⁴Eu were present in addition to ¹³⁷Cs, the ¹⁵⁴Eu gamma rays would also contribute counts to the Cs window, and the amount of ¹³⁷Cs would be over-estimated. Worse, the ¹⁵⁴Eu would most likely go undetected because the gamma rays listed in the table above would not be counted in the potassium, uranium, or thorium windows.

background subtraction coefficients cannot be made because there are no calibration

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The adaptation of the NURE spectral stripping technique is ineffective for Hanford logging because multiple man-made gamma-ray sources produce backgrounds in the cesium window that cannot be calculated. Measurements from which to derive

standards that contain the necessary man-made radionuclides, both individually and in combination. Evaluation of gamma energy spectral peaks instead of window counts is not feasible because the NaI(Tl) detectors have such poor energy resolution that the spectra from contaminated zones contain numerous obscure and overlapping peaks.

These considerations led to a re-evaluation of the overall analytical approach. Specific radionuclides have been identified by the baseline characterization program, and concentrations are known. Therefore, the primary goal of the RAS is to detect *changes* in radioactivity levels. Decreases in concentrations consistent with radioactive decay are expected, but contaminant migration may be indicated by either increases or decreases in radioactivity levels that cannot be explained by decay.

A revised analytical approach was developed which is still based on spectral windows. The four spectral windows for Cs, K, U and T are retained (although the energy ranges have been modified) and four additional windows are defined to cover the entire energy range of the detector. Counts are collected for all eight windows as well as total counts. Table 2 identifies the energy range of each spectral window. Channel ranges for each detector are also listed.

Table 2. RAS Energy Windows

	Energy range		Channel range			
	Window	keV	Large (L)	Medium (M)	Small (S)	
1	Lithology	0 - 570	0 - 52	0 - 49	0 - 52	
2	Cesium	570 - 740	53 - 68	50 - 64	53 - 67	
3	Midrange	740 - 940	69 - 86	65 - 80	68 - 84	
4	Protactinium	940 - 1060	87 - 97	81 – 90	85 – 95	
5	Cobalt	1060 - 1390	98 - 126	91 – 118	96 – 123	
6	Potassium	1390 - 1600	127 - 145	119 – 135	124 - 140	
7	Uranium	1600 - 2400	146 - 214	136 - 200	141 - 206	
8	Thorium	2400 - 2800	215 - 255	201 - 255	207 - 255	

The potassium, uranium and thorium windows are defined to track naturally occurring radionuclides. The cesium and cobalt windows are defined to track specific man-made radionuclides. A protactinium window captures counts from protactinium-234m (^{234m}Pa), an early daughter in the ²³⁸U decay series, which quickly reaches secular equilibrium with the parent ²³⁸U. Because ^{234m}Pa has a relatively low gamma yield, its characteristic gamma rays are not detected from natural uranium at typical concentration levels. Hence, the presence of gamma rays associated with this radionuclide can be taken as an indication of purified uranium in which the decay series has been perturbed by chemical processing, so that the concentration of ²³⁸U is high, but the concentrations of the uranium decay progenies below ^{234m}Pa are extremely low. Finally, the lithology and midrange windows fill the gap between the other windows. The sum of counts in the eight individual windows should be equal to the total counts.

To a first approximation, changes in count rates can be identified by simply comparing plots of successive log runs. When necessary, decay corrections can be made using

radionuclide identification from the baseline data. Areas of possible contaminant migration can be identified by changes in count rate over a depth interval or changes in the depth over which anomalous activity occurs.

Like all radiation measurements, RAS data are subject to random fluctuations associated with the radioactive decay process. Therefore, it will be necessary to determine if observed differences in count rates are statistically significant. This follows a method described in Knoll (2000).

 N_1 and N_2 designate two individual measurements taken at different times (assume appropriate decay corrections have been made to correct both count rates to a common time). Both are taken to be estimates of the mean value of a Gaussian distribution at the time of measurement. The estimate for the standard deviation is equivalent to the square root of the counts.

$$\sigma = \sqrt{N} \tag{2}$$

The count rates, R_1 and R_2 , are determined by dividing the counts by the live time. The count rate also represents a Gaussian (normal) distribution, since R = N/T. The estimate of the standard deviation for the count rate is:

$$\sigma = \frac{\sqrt{N}}{T} = \frac{\sqrt{RT}}{T} = \sqrt{\frac{R}{T}} \tag{3}$$

The difference in count rates between the measurements should also follow a Gaussian distribution.

If there is no actual difference in the two counts, then the true mean values for R_1 and R_2 are the same and:

$$\sigma_{\Delta R} = \sqrt{\sigma_{R_1}^2 + \sigma_{R_2}^2} \tag{4}$$

We can define a critical level, L_1 , so that the probability of false positives is minimal. For the purpose of this analysis, we will accept a 5% chance for a false positive result. For a one-tailed normal distribution, there is a 95% probability that a random sample of R_2 will lie below the mean + 1.645 σ when R_2 and R_1 are taken from the same distribution.

Also,
$$\sigma_{R1} \approx \sigma_{R2}$$
, so that: $\sigma_{\Delta R} = \sqrt{\sigma_{R_2}^2 + \sigma_{R_1}^2} = \sqrt{2} \times \sigma_{R_1}$ (5)

Therefore:
$$L_1 = R_1 + 1.645 \times \sqrt{2} \times \sigma_{R_1} = R_1 + 2.326 \times \sigma_{R_2}$$
 (6)

In the case where a real difference in activity exists, the true mean value for ΔR is >0, and we can define a minimum limit for R_2 for which the probability of false negatives is

minimal. If $R_2 = L_1$, the false negative rate will be 50 %, because a Gaussian distribution is symmetric about its mean. To ensure that 95 % of the values in the R_2 distribution lie above L_1 , we define L_2 so that:

$$L_2 = L_1 + 1.645 \times \sigma_{\Lambda R} \tag{7}$$

also,
$$\sigma_{R2} \ge \sigma_{R1}$$
, so that: $\sigma_{\Delta R} = \sqrt{\sigma_{R_2}^2 + \sigma_{R_2}^2} \le \sqrt{2} \times \sigma_{R_2}$ (8)

Therefore:
$$L_2 = R_1 + 2.326 \times \sigma_{R_1} + 2.326 \times \sigma_{R_2}$$
 (9)

L₂ defines the level above which there is a 95% probability that the count rates are different.

For radiation measurements,
$$\sigma_N \approx \sqrt{N}$$
: $\sigma_R = \sqrt{\frac{R}{T}}$ (10)

This leads to definition of two limit values based on count rates, which can be used to compare successive monitoring runs.

$$L_1 = R_1 + 2.326 \times \sqrt{\frac{R_1}{T_1}} \tag{11}$$

$$L_2 = R_1 + 2.326 \times \sqrt{\frac{R_1}{T_1}} + 2.326 \times \sqrt{\frac{R_2}{T_2}}$$
 (12)

Data evaluation consists of comparing the second count rate to these limits:

 $R_2 \le L_1$ \Rightarrow no significant difference (95%)

 $L_1 < R_2 < L_2 \implies \text{ambiguous}$

 $R_2 \ge L_2$ \Rightarrow significant difference (95%)

Values for R_1 , L_1 and L_2 should be corrected for decay for the time at which R_2 is measured. These values may be calculated for specific windows or for total counts. When the second count rate lies between L_1 and L_2 , it is likely that a difference in count rates exists, but at a lower confidence interval. In this case, the ambiguity can possibly be resolved by comparing with a previous count rate over a longer time interval, comparing changes in a different window, or by calculating new limits based on a lower degree of confidence.

The above equations are relatively simple and can be implemented in a Microsoft EXCEL® spreadsheet. Data from the RAS consists of window counts as a function of depth written as text files. These files can be imported to the spreadsheet and plotted or

analyzed in greater detail when necessary. If needed, decay corrections can be easily calculated and applied.